# PATENT COOPERATION TREATY

# **PCT**

## INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

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Applicant's or agent's file reference 63446A	FOR FURTHER ACTION	See Form PCT/IPEA/416
International application No. PCT/US2004/030706	International filing date (day/mont 17.09.2004	hlyear) Priority date (day/monthlyear) 19.09.2003
International Patent Classification (IPC) or na C09J123/08, C08F210/02, C08L23/0		
Applicant DOW GLOBAL TECHNOLOGIES IN	NC. et al.	
This report is the international pre- Authority under Article 35 and trans		ablished by this International Preliminary Examining ing to Article 36.
2. This REPORT consists of a total of	of 6 sheets, including this cover	sheet.
3. This report is also accompanied b	v ANNEXES comprising:	
a. 🗵 sent to the applicant and to		al of 16 sheets as follows:
☐ sheets of the description	on, claims and/or drawings which	h have been amended and are the basis of this report is Authority (see Rule 70.16 and Section 607 of the
sheets which supersed beyond the disclosure Supplemental Box.	le earlier sheets, but which this in the international application a	Authority considers contain an amendment that goes as filed, as indicated in item 4 of Box No. I and the
sequence listing and/or tab	ureau only) a total of (indicate ty les related thereto, in computer Listing (see Section 802 of the	rpe and number of electronic carrier(s)) , containing a readable form only, as indicated in the Supplemental Administrative Instructions).
4. This report contains indications re	lating to the following items:	
☑ Box No. I Basis of the opir	nion	
☑ Box No. II Priority		
	ent of opinion with regard to nov	velty, inventive step and industrial applicability
☐ Box No. IV Lack of unity of	· · · · · · · · · · · · · · · · · · ·	The state of the s
☐ Box No. V Reasoned state		gard to novelty, inventive step or industrial ing such statement
☐ Box No. VI Certain docume		
☐ Box No. VII Certain defects i	in the international application	
⊠-Box-No:-VIIIGertain-observa	tions-on-the-international-applica	ation
Date of submission of the demand	Date of	completion of this report
21.04.2005	15.12.	2005
Name and mailing address of the international preliminary examining authority:	al Authoriz	zed Officer
European Patent Office D-80298 Munich	t, S	
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# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No. PCT/US2004/030706

	Box No. I Basis of the report	
	<del></del>	
1.	With regard to the language, this filed, unless otherwise indicated	s report is based on the international application in the language in which it was under this item.
	which is the language of a tr international search (und publication of the interna	tional application (under Rule 12.4)
	☐ international preliminary	examination (under Rules 55.2 and/or 55.3)
2.	With regard to the elements* of have been furnished to the receireport as "originally filed" and are	the international application, this report is based on (replacement sheets which ving Office in response to an invitation under Article 14 are referred to in this e not annexed to this report):
	Description, Pages	
	1-6, 8-32, 34, 36-40, 42, 45	as originally filed
	7, 7a, 33, 35, 41, 43, 44, 46, 46a, 46b, 47	received on 25.04.2005 with letter of 21.04.2005
	Claims, Numbers	
	1-20	received on 25.04.2005 with letter of 21.04.2005
	☐ a sequence listing and/or an	y related table(s) - see Supplemental Box Relating to Sequence Listing
3.	☐ The amendments have resu	ulted in the cancellation of:
	☐ the description, pages	•
	☐ the claims, Nos.	
	☐ the drawings, sheets/figs☐ the sequence listing (spe	
	any table(s) related to se	
4.		ished as if (some of) the amendments annexed to this report and listed below have been considered to go beyond the disclosure as filed, as indicated in the ).
	<ul><li>☐ the description, pages</li><li>☐ the claims, Nos.</li></ul>	
	☐ the drawings, sheets/figs	
	☐ the sequence listing (spe ☐ any table(s) related to se	
	-* -If-item-4-applies, so	ome or all of these sheets may be marked "superseded."

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No. PCT/US2004/030706

_	Box	x No. II Priority	
1.	Ø	This report has been established prescribed time limit the requeste	as if no priority had been claimed due to the failure to furnish within the
		☐ copy of the earlier application	whose priority has been claimed (Rule 66.7(a)).
		$\square$ translation of the earlier application	ation whose priority has been claimed (Rule 66.7(b)).
2.			as if no priority had been claimed due to the fact that the priority claim has us for the purposes of this report, the international filing date indicated vant date.
3.	Add	ditional observations, if necessary:	
_			
_		x No. III Non-establishment of on the plicability	ppinion with regard to novelty, inventive step and industrial
1.	The obv	e questions whether the claimed invious), or to be industrially applicab	rention appears to be novel, to involve an inventive step (to be non- e have not been examined in respect of:
		the entire international application	,
	$\boxtimes$	claims Nos. 1-20	
		because:	·.
		the said international application, not require an international prelim	or the said claims Nos. relate to the following subject matter which does inary examination (specify):
	☒	the description, claims or drawing unclear that no meaningful opinio	s <i>(indicate particular elements below)</i> or said claims Nos. 1-20 are son could be formed <i>(specify)</i> :
		see separate sheet	
		the claims, or said claims Nos. ar could be formed.	e so inadequately supported by the description that no meaningful opinion
		no international search report has	been established for the said claims Nos.
		the nucleotide and/or amino acide C of the Administrative Instruction	sequence listing does not comply with the standard provided for in Annex is in that:
		the written form	has not been furnished
		[	does not comply with the standard
		_the_computer_readable_form[	has_not_been_furnished
		[	does not comply with the standard
		the tables related to the nucleotid not comply with the technical requ	e and/or amino acid sequence listing, if in computer readable form only, do uirements provided for in Annex C- <i>bis</i> of the Administrative Instructions.
		See separate sheet for further de	tails

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No. PCT/US2004/030706

## Box No. VIII Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

see separate sheet

PCT/US2004/030706

#### ad III:

1. For the time being, the subject-matter as presently defined in the claims is considered to be unclear (see item VIII below) so that no meaningful substantive examination could be carried out.

#### ad VIII:

- 1. The Applicants defined in independent claims 1 and 9 the ratio of the high weight average molecular weight fraction to the low weight average molecular weight fraction M<sub>wH</sub>/M<sub>wL</sub> being from "about 1 to about 20", in particular, the term "about" has been introduced in order to overcome the objection made in the Written Opinion under item VIII 1.3. Applicants argued that "about" should provide for numerical values slightly greater than 1 and according to the Applicants it should be understood from the claim language that "about 1" refers to "numbers slightly greater than 1".
- 2. It is considered that the expression "about 1" does not only refer to "numbers slightly greater than 1" but also to "numbers slightly smaller than 1". That is why the term "about" in general is considered to be an unclear term (PCT Guidelines, C-III, 4.5a).
  In any case, "about 1" is to be read as "1", or alternatively, "1" is to be read with error margins, ie "about 1" so that the introduction of the word "about" did not produce any difference having regard to the ratio range.
- 3. Therefore, for the time being, the subject-matter of the claims is considered to be unclear, as it has not been made clear that in fact the high weight average molecular weight fraction has a higher molecular weight than the low weight average-molecular-weight-fraction.
- 4. For the regional phase, the Applicant is asked to delete the term "about" in the claims, and to replace the lower limit for the range of ratios  $M_{wH}/M_{wL}$  "about 1" by the number "1.5" or "2" (as is indicated for instance on page 11, lines 16-17) in order to make clear that in fact the high weight average molecular weight fraction has a higher molecular weight than the low weight average molecular weight

## INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

International application No.

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fraction.

5. Please note, that this feature is considered as an essential feature, distinguishing the the present claimed subject-matter from the disclosure of the cited prior art. The Applicants mentioned this feature at first place as the distinguishing feature from prior art D1 to D3 (cf. letter of 21/04/2005 page 2, last paragraph; page 3, last paragraph; and page 4, fourth paragraph). A feature essential for defining the invention has to be defined clearly (Article 6 PCT).

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The ethylene alpha olefin interpolymer of the present invention has a Brookfield Viscosity (measured at 300°F/149°C) of from about 500 (0.5), preferably about 1,000 (1.0), and more preferably from about 1,500 (1.5) up to about 7,000 cP (7.0 Pa·s), preferably to about 6,000 cP (6.0 Pa·s), more preferably up to about 5,000 cP (5.0 Pa·s). The interpolymer may have a Brookfield Viscosity (measured at 300°F/149°C) from 500 to 9000 cP (0.5 to 9.0 Pa·s).

The ethylene alpha olefin interpolymer of the present invention when mixed with a tackifier results in an adhesive composition having a Brookfield Viscosity (measured at 350°F/177°C) of from about 400 (0.4), preferably about 500 (0.5) and more preferably from about 750 (0.75) up to about 2,000 cP (2.0 Pa·s), preferably to about 1,400 cP (1.4 Pa·s), more preferably up to about 1,200 cP (1.2 Pa·s).

The ethylene alpha olefin interpolymer of the present invention when mixed with a tackifier results in an adhesive composition having a Peel Adhesion Failure Temperature (PAFT) of greater than or equal to 110°F (43.3°C), preferably greater than or equal to 115°F (46.1°C), more preferably greater than or equal to 120°F (48.8°C).

The ethylene alpha olefin interpolymer of the present invention when mixed with a tackifier results in an adhesive composition having a Shear Adhesion Failure Temperature (SAFT) of greater than or equal to 140°F (60°C), greater than or equal to 150°F (65.5°C), more preferably greater than or equal to 170°F (76.7°C).

The ethylene alpha olefin interpolymer of the present invention when mixed with a tackifier results in an adhesive composition which exhibits 100% paper tear from 77 to 140°F (25° to 60°C), preferably 100% paper tear from 35 to 140°F (1.7° to 60°C), and most preferably 100% paper tear from 0 to 140°F (negative 17.7°C to 60°C).

The resulting adhesive compositions noted above, suitably serve as hot melt adhesives when appropriately formulated, for various end applications in which such HMAs typically are employed.

Another embodiment of the invention provides a process of making an ethylene alpha olefin interpolymer, comprising (a) contacting one or more olefinic monomers in the presence of at least two catalysts; and (b) effectuating the polymerization of the olefinic monomers in one or more reactors to obtain an olefin polymer, wherein each

catalyst has the ability to incorporate a different amount of comonomer in the polymer, and/or wherein each catalyst is capable of producing a polymer with substantially



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tackifier to modify viscosity and improve the tack properties of an adhesive composition.

A dispersant can also be added to these compositions. The dispersant can be a chemical, which may, by itself, cause the composition to be dispersed from the surface to which it has been applied, for example, under aqueous conditions. The dispersant may also be an agent which when chemically modified, causes the composition to be dispersed from the surface to which it has been applied. As known to those skilled in the art, examples of these dispersants include surfactants, emulsifying agents, and various cationic, anionic or nonionic dispersants. Compounds such as amines, amides and their derivatives are examples of cationic dispersants. Soaps, acids, esters and alcohols are among the known anionic dispersants. The addition of a dispersant may affect the recyclability of products to which a hot-melt adhesive may have been applied.

The surfactants can be chosen from a variety of known surface-active agents. These can include nonionic compounds such as ethoxylates available from commercial suppliers. Examples include alcohol ethoxylates, alkylamine ethoxylates, alkylphenol ethyoxylates, octylphenol ethoxylates and the like. Other surfactants, such as a number of fatty acid esters may be employed; for example, but not limited to, glycerol esters, polyethyleneglycol esters and sorbitan esters.

### 20 Tackifiers

In order to formulate hot melt adhesives from the polymers of the present invention, the addition of tackifier is desirable to allow for bonding prior to solidifying or setting of the adhesive. An example of this is in high-speed cereal box sealing operations where the overlapping flaps of the box need to adhere to one another while the hot melt adhesive solidifies.

Such tackifying resins include aliphatic, cycloaliphatic and aromatic hydrocarbons and modified hydrocarbons and hydrogenated versions; terpenes and modified terpenes and hydrogenated versions; and rosins and rosins derivatives and hydrogenated versions; and mixtures thereof. These tackifying resins have a ring and ball softening point from 70°C to 150°C, and will typically have a viscosity at 350°F (177°C), as measured using a Brookfield viscometer, of no more than 2000 centipoise (2.0 Pa·s).

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Another exemplary tackifier, Piccotac 115, has a viscosity at 350°F (177°C) of about 1600 centipoise (1.6 Pa·s). Other typical tackifiers have viscosities at 350°F (177°C) of much less than 1600 centipoise (1.6 Pa·s), for instance, from 50 to 300 centipoise (0.05 to 0.3 Pa·s).

Exemplary aliphatic resins include those available under the trade designations Eastotac<sup>TM</sup>, Escorez<sup>TM</sup>, Piccotac<sup>TM</sup>, Mercures<sup>TM</sup>, Wingtack<sup>TM</sup>, Hi-Rez<sup>TM</sup>, Quintone<sup>TM</sup>, Tackirol<sup>TM</sup>, etc. Exemplary polyterpene resins include those available under the trade designations Nirez<sup>TM</sup>, Piccolyte<sup>TM</sup>, Wingtack<sup>TM</sup>, Zonarez<sup>TM</sup>, etc. Exemplary hydrogenated resins include those available under the trade designations Escorez<sup>TM</sup>, Arkon<sup>TM</sup>, Clearon<sup>TM</sup>, etc. Exemplary mixed aliphatic-aromatic resins include those available under the trade designations Escorez<sup>TM</sup>, Regalite<sup>TM</sup>, Hercures<sup>TM</sup>, AR<sup>TM</sup>, Imprez<sup>TM</sup>, Norsolene<sup>TM</sup> M, Marukarez<sup>TM</sup>, Arkon<sup>TM</sup> M, Quintone<sup>TM</sup>, etc. Other tackifiers may be employed, provided they are compatible with the homogeneous linear or substantially linear ethylene/alpha.-olefin interpolymer.

Although the present invention has been described with a certain degree of particularity, it is to be understood that the examples below are merely for purposes of illustrating the present invention, the scope of the present invention is not intended to be defined by the claims.

## PREPARATION OF EXAMPLES

Unless otherwise stated, the following test methods are employed and percentages or parts are by weight.

Density is measured in accordance with ASTM D-792. The samples are annealed at ambient conditions for 24 hours before the measurement is taken.

Comonomer content of the invention polymer is determined by Nuclear Magnetic Resonance (NMR) analysis. The analysis sample is prepared by adding about 3g of a 50/50 mixture of tetrachloroethane-d<sup>2</sup>/ortho-dichlorobenzene (to which sufficient chromium acetylacetonate is added so the mixture is 0.025M in the chromium compound) to a 0.4g sample of the polymer in a 10mm NMR tube. Samples are dissolved and homogenized in the tube by heating it and contents to 150°C/302°F. Data is collected using a Varian Unity Plus 400MHz NMR spectrometer, corresponding to a <sup>13</sup>C resonance frequency

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The ethylene or ethylene / hydrogen mixture combined with the solvent / comonomer stream at ambient temperature. The temperature of the solvent/monomer as it enters the reactor was controlled with two heat exchangers. This stream enters the bottom of the 1 gallon continuously stirred tank reactor.

In an inert atmosphere box, a solution of the transition metal compounds was prepared by mixing the appropriate volumes of concentrated solutions of each of the two components with solvent to provide the final catalyst solution of known concentration and composition. This solution was transferred under nitrogen to a pressure vessel attached to a high-pressure metering pump for transport to the polymerization reactor.

In the same inert atmosphere box, solutions of the primary cocatalyst, methylbis(hydrogenatedtallowalkyl) ammonium tetrakis (pentafluorophenyl)borate and the secondary cocatalyst, MMAO Type 3A, were prepared in solvent and transferred to separate pressure vessels as described for the catalyst solution. The ratio of Al to transition metal and B to transition metal was established by controlling the volumetric flow output if the individual metering pumps to attain the molar ratios in the polymerization reactor as presented in Table 2. The multiple component catalyst system and its solvent flush also enter the reactor at the bottom but through a different port than the monomer stream.

Polymerization was stopped with the addition of water into the reactor product line after the meter measuring the solution density. The reactor effluent stream then entered a post reactor heater that provides additional energy for the solvent removal flash. This flash occurs as the effluent exits the post reactor heater and the pressure is dropped from 475 psig (3,275 kPa) down to 10 at the reactor pressure control valve.

This flashed polymer entered a hot oil jacketed devolatilizer. Approximately 90 % of the volatiles were removed from the polymer in the devolatilizer. The volatiles exit the top of the devolatilizer. The remaining stream is condensed with a chilled water jacketed exchanger and then enters a glycol-jacket solvent / ethylene separation vessel. Solvent is removed from the bottom of the vessel and ethylene vents from the top. The ethylene stream is measured with a Micro-Motion mass flow meter. This measurement of unreacted ethylene was used to calculate the ethylene conversion. The



Table 3 - Ethylene/α-Olefin Interpolymers Preparation Conditions

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1	,"		7		_				_		_				_		7		_				$\neg$
74/ H.				13/3		13/3		13/3		13/3		13/90		13/90		13/90		13/8	,	13/90		3/8	
Mole	Ratio	Catalyst		1:1	,	1:3		1:3		1:3		1:1		1:20		1:20		∷		1:10		2:1	
Catalysts				CATS-1/2	,	<b>CATS-1/2</b>		<b>CATS-1/2</b>		<b>CATS 1/2</b>		CATS 1/3		CATS 1/3		<b>CATS 1/3</b>		CATS-1/4		CATS 1/3	,	CATS 2/4	
MMAO /Tr	Molar Ratio			10.07		6.01	•	6.04		5.96		66.5		5.83		5.93		4.95		4.91		4.95	
B%Tr <sup>c</sup>	Molar			1.21		1.47		1.51		1.37		1.47		1.48		1.49		1.06		1.08		1.08	
C2	Conversion	%		74.68		69'68		20'32		80.15		90.46	•	90.13	•	90.03		90.40		90.44	•	90.35	
Hydrogen				174.48		111.75		113.80		150.35		71.76		06'69		47.98		83.20		13.45	,	121.97	
Octene	Flow	lb/hr	(kg/hr)	1.25	(0.57)	0.86	(0.39)	92.0	(0.34)	0.85	(0.39)	1.03	(0.47)	1.06	(0.48)	1.17	(0.53)	1.00	(0.45)	1.60	(0.73)	0.73	(0.33)
Ethylene	Flow	lb/hr	(kg/hr)	2.68	(1.22)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)	2.65	(1.20)
Solvent	Flow	lb/hr	(kg/hr)	25.20	(11.43)	25.76	(11.68)	25.80	(11.70)	25.77	(11.69)	20.87	(9.46)	20.81	(9.44)	20.78	(9.42)	25.51	(11.57)	25.20	(11.43)	25.60	(11.61)
Reactor	Temp	ပ္စ		150.32		150.50		150.38		149.88		129.73		130.03		119.13		149.65		120.28		150.20	
五 五						7		3		4	-	5		9		7	_	8		6		2	

The primary cocatalyst for all polymerizations was Armeenium Borate, [methylbis(hydrogenatedtallowalkyl) ammonium tetrakis (pentafluorophenyl)borate prepared as in U.S. Patent # 5,919,983, Ex. 2, the entire disclosure of which patent is incorporated herein by reference.

<sup>b</sup> The secondary cocatalyst for all polymerizations was a modified methylalumoxane (MMAO) available from Akzo Nobel as MMAO-3A (CAS# 146905-79-10).

For Examples 1-4, 8 and 10 the term Tr refers to the total titanium content of the mixed catalyst system. For Examples 5-7 and 9 the term Tr refers to the Zr content only of the mixed catalyst system.

For Examples 1-4 and 8 it can be noted that the  $r_1^H/r_1^L$  ratio exceeds unity and, surprisingly (see Table 5), properties of formulations made from such interpolymers are quite good and comparable to those from Examples 5-7 and 9-10. 9

Table 4- Properties of Ethylene/1-Octene Interpolymers

_			,		<del>,</del>						
Te3	ව්										55.1
Te2	<u>ල</u> .	55.0	73.4	77.1	78.2	52.1		81.1	54.2		82.3
T, 1	ව	97.1	8.66	101.2	102.7	91.1	94.7	97.1	93.6	100.8	95.0
%	Cryst	33	39	42	43	41	43	45	45	47	. 94
Heat of	Fusion (J/g)	96.2	113.3	121.4	125.9	120.7	125.8	130.9	130.2	136.7	134.7
T	ဥ	111.1	114.6	115.6	116.1						110.6
T	<u>ව</u>	107.0	110.3	111.4	112.3		107.2	110.2	103.3	113.7	105.6
Tai	ତ୍ର .	81.2	86.3	89.4	90.0	103.3	95.1	93.7	<b>2.96</b>	93.1	93.3
Drop	Point	113.3	116.9	117.8	118.4	109.6	112.1	113.5	109.6	116.1	114.6
Wol%	Com.	7.10	5.81	5.30	5.23	5.34	5.50	5.64	5.10	5.50	6.30
Wt%	Com	23.40	19.80	18.30	18.10	18.40	18.90	19.30	17.70	18.80	18.30
Mw/Mn		2.29	2.23	2.16	2.15	4.15	5.77	8.05	2.39	15.04	2.74
M		4,180	5,030	5,220	5,060	2,700	2,080	1,590	4,610	1,130	3,940
Mw		0,570	11,200	11,300	006'01	11,200	12,000	12,800	11,000	17,000	10,800
		0.8941	0.9040	0.9083	0.9092	0.9091	0.9089	0.9052	0.9086	0.9067	0.9084
Ex# Viscosity	@ 300 F cP (Pa·s)	1,600 (1.600)	2,879 (2,879)	2,859 (2.8 <u>5</u> 9)	2,744 (2,7,44)	2,804 (2.804)	2,889 (2,889)	2,684 (2.684)	3,047 (3.047)	3,113	2,855 (2.855)
Ex#		1	2	3	4	2	9	7	∞	6	10

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Table 5 - Properties of Hot Melt Adhesives Made From Ethylene/Octene Interpolymers of the Present Invention

!_																									•						
	Viscosity (@_550, F (177 °C) cP (Pars)	· · · · · · · · · · · · · · · · · · ·		1,115	(1.115)	1,050	(1.050)	. 056	(0.950)	1,060	(1.060)	935	(0.935)	820	(0.820)	1,080	(1.080)	086	(0.980)	099	(0.660)	270	(0.570)	200	(0.500)	470	(0.470)	1,050	(1.050)	096	(0.960)
	%F (C)			202	(96.1)	203	(95.0)	201	(63.9)	211	(99.4)	208	(97.8)	208	(87.8)	215	(102)	212	(100)	211	(99.4)	215	(102)	213	(101)	211	(99.4)	203	(0.56)	202	(94.4)
2 1	(%)			110	(43.3)	119	(48.3)	128	(53.3)	110	(43.3)	118	(47.8)	131	(55.0)	110	(43.3)	132	(55.6)	156	(68.9)	120	(48.9)	122	(50,0)	132	(55.6)	111	(43.9)	115	(46.1)
ŀ	•	140 F	(00.00	<u> </u>		. 00		100		75		. 001	,	100		100	. *	100		100		75		100		100		20		8	
	:	120 F	(48.9°C)	9		6		100		100		100		92	1	100		100		100		100		100		100		100		<u>8</u>	
.0)	raper rear (%)	77 °F	(2.0)						,	100		100		8		100	•	100		100		8		은 은		8		100		01	
-		35 F	() () ()	23		 O		0		100		90		0		20		. 25		0		જ		ઇ		0		<u>8</u>		23	
	•	0 作 5 8 6 5 6 5 6 5 6 5 6 5 6 5 6 5 6 5 6 5	(7-8-/1-)	0		0		0		0		0		0		• •		0		0		0		0		0		0		<u> </u>	
Breezes	5637 (wt%)		8	7.7		27	,	32		22		27		32		22		27		32		22		27		32		22	,,	72	
Dolumor	(wt%)		2	<b>x</b>		73		89		28		23		89		8		73		86		%		25		8		%		55	
Polemer	Ex#	•	•			_				7		2		7		m		m		<u>س</u>		4		4		4		ς.		٠ دى	_



•	Viscosity @ 350 °F (177 °C)	св. (Ряз)			098	1 000	(1.000)	945	(0.945)	850	(0.850)	925	(0.925)	. 840	(0.840)	755	(0,755)	1300	(1.300)	1205	(1.205)	1100	(1.100)	· 1035	(1.035)	1140	(1.140)	1070	(1:070)	930	(0.930)
	SAFT %	(၁)			200	203	(95.0)	202	(94.4)	200	(93.3)	209	(68.3)	202	(97.2)	205	(96.1)	214	(101)	208	(97.8)	207	(2,76)	207	(57.2)	212	(100)	210	(6.86)	208	(97.8)
	PAFT F	(၁)	•		118	104	(40)	115	(46.1)	124	(51.1)	95	(35)	109	(42.8)	127	(52.8)	06	(32.2)	109	(42.8)	126	(52.2)	128	(53.3)	96	(32.2)	06	(32.2)	8	(32.2)
			140 F	(60.0°C)	100	18	3	100		100		20	•	75		100		100		100		100		001		0	-	0		75 .	
Table 5 - continued	*		120 F	(48.9°C)	100	100	3	100		100		20		100		100		100	•	100		100		100		20	,	100		100	
Sable 5-	Paper Tear (%) *		77 ºF	(25°C)	100	100	3	100		100		NW		100		100		MM		MM		MM		NM	•	MN		MM		MA	•
	Pap		35 °F	(1.7°C)	0	95	3	0		0		25		25		0		100	*	20		0		.0	-	100	•	100		100	
			0 F	(-17.8°C)	0	٥	>	. 0		0		. 25		0		0		0		0		0		0		100	•	001		75	
	Escorez 5637 (wt%)				. 32	22		12	•	32		22		27		32		. 11.		22		27		32		17		22	,	27	
	Polymer (wt%)				89	2,8	2	73		89		78		73		89		83		78		73		89		83		78	,	73	

46a

Polymer Ex#

46b

	į.	•	•												
Viscosity @ 350 Tr	• •	C. C.	(Pas)	11		810	(0.810)	1175	(1.175)	1115	(1.115)	1040	(1.040)	920	(0.920)
SAFT	딾	<u>်</u>		<u></u>		807	(87.8)	202	(96.1)	204	(92.6)	202	(94.4)	203	(95.0
PAFT	দ	ဉ				111	(43.9)	105	(40.0)	112	(44.4)	126	(52.2)	131	(55.0)
-				140 F	(2°0.09)	100		· 001		100		100		100	
* (				120 F	(48.9°C)	100	•	100		100		100		100	
per Tear (%				77 F	(25°C)	MIN		MM		MM		MN		ME	
Pa				35 F	(1.7°C)	100		100	•	100		100		0	
_				0ች	(-17.8°C)	0		0		0		0		0	
Escorez	5637 (wt%)				;	32		11		22		27		32	
Polymer	(wt%)	•				89		83		78		73		89	
Polymer	#X					. 6		. 01		10		10		10	
	Polymer Escorez Paper Tear (%) * PAFT SAFT Viscosity @ 350 F	Polymer   Escorez   Paper Tear (%) *   PAFT   SAFT   Viscosity @ 350 °F : (177 °C) - (	Polymer   Escorez   Paper Tear (%) *   PAFT   SAFT   Viscosity @ 350 F.   (wt%)   5637 (wt%)   ("C")   ("C")	Polymer   Escorez   Paper Tear (%) * PAFT   SAFT   Viscosity @ 350 F   (177 °C) - cP   (°C)   (°C)	Polymer         Escorez         Paper Tear (%) *         PAFT         SAFT         Viscosity @ 350 F           (wt%)         5637 (wt%)         °C         °F         °F         (177 °C) -           (°C)         (°C)         (°C)         °C         (°C) -           (°C)         (°C)         (°C)         (°C)           (°C)         (°C)         (°C)         (°C)	Polymer Escorez Paper Tear (%) * PAFT SAFT Viscosity @ 350 F (177 °C) - C (°C) (°C) (°C) (°C) (°C) (°C) (°C) (°	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) *         Paper Tear (%) *	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         PAFT (%) *         PAFT (%) *         PAFT (177 °C) (%) *           (wt%)         5637 (wt%)         (%) <td< td=""><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         Paper Tear (%) **         Paper Tear (%) **         PAFT (%)         PAFT (%)         Viscosity (@) 350 °F (177 °C) (177 °C)           (wt%)         5637 (wt%)         (%)         <t< td=""><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td><td>  Polymer   Escorez   Paper Tear (%) *   PAFT   SAFT   Viscosity @ 350 °F.     (wt%)   5637 (wt%)   S637 (wt%)   S637 (wt%)     (wt%)   5637 (wt%)   S637 (wt%)  </td><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td><td>Polymer         Escorez         Paper Tear (%) **         PAFT         SAFT         Viscosity @ 350 °F.           (wt%)         5637 (wt%)         0°F         35 °F         170 °F         120 °F         140 °F         77 °C)         (°C)         &lt;</td><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td></t<></td></td<>	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         Paper Tear (%) **         Paper Tear (%) **         PAFT (%)         PAFT (%)         Viscosity (@) 350 °F (177 °C) (177 °C)           (wt%)         5637 (wt%)         (%) <t< td=""><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td><td>  Polymer   Escorez   Paper Tear (%) *   PAFT   SAFT   Viscosity @ 350 °F.     (wt%)   5637 (wt%)   S637 (wt%)   S637 (wt%)     (wt%)   5637 (wt%)   S637 (wt%)  </td><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td><td>Polymer         Escorez         Paper Tear (%) **         PAFT         SAFT         Viscosity @ 350 °F.           (wt%)         5637 (wt%)         0°F         35 °F         170 °F         120 °F         140 °F         77 °C)         (°C)         &lt;</td><td>Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P</td></t<>	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P	Polymer   Escorez   Paper Tear (%) *   PAFT   SAFT   Viscosity @ 350 °F.     (wt%)   5637 (wt%)   S637 (wt%)   S637 (wt%)     (wt%)   5637 (wt%)   S637 (wt%)	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P	Polymer         Escorez         Paper Tear (%) **         PAFT         SAFT         Viscosity @ 350 °F.           (wt%)         5637 (wt%)         0°F         35 °F         170 °F         120 °F         140 °F         77 °C)         (°C)         <	Polymer (wt%)         Escorez (wt%)         Paper Tear (%) **         P

\* NM = not measured

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Table 6 - Properties of Commercial Hot Melt Adhesives

Comp Ex	Commercial Name	Viscosity @ 350°F		Par	Paper Tear (%)	. (9		PAFT F	SAFT
	,	(177°C) cP	•	-				ව්	ව
		(Pa·s)			•				
	•			35 F	77 T	120 F	140 또		
			(-17.8°C)	(1.7°C)	(25°C)	(48.9°C)	(2 <sub>0</sub> 9)		
-	ADVANTRA HL.	860	901	100	100	001	100	142	198
	9250	(0.860)						(61.1)	(92.2)
2	ADVANTRA HL-	750	0	100	100	100	100	151	192
	9256	(0.750)						(66.1)	(88.9)
3	HL-7268	096	:	:	100	100	100	144	192
		(0.960)	6				•	(62,2)	(88.9)
4	HL-2835	1,070	901	100	100	100	100	126	153
		(1.070)	,					(52.2)	(67.2)
5	80-8488	1,080	ŀ	001	100	100	100	150	176
	al	(080.1)						(65.6)	(80.0)
9	80-8368	026	-	100	100	100	100	142	190
		(0.970)						(61.1)	(87.8)

**20** 

#### CLAIMS.

#### We claim:

- 5 1. A composition comprising an ethylene interpolymer having
  - i) a number average molecular weight (Mn) from 1,000 to 9,000; and
  - ii) a Brookfield Viscosity (measured at 149°C/300°F) from 500 to 9,000 cP (0.5 to 9.0 Pa·s);

and one or more tackifiers, added in an amount from 15 to 40 percent by weight (based on the combined weight of interpolymer and tackifier), and wherein the composition has:

- a) a Brookfield Viscosity (measured at 177°C/350°F) from 400 to 2,000 cP (0.4 to 2.0 Pa·s);
- b) a Peel Adhesion Failure Temperature (PAFT) of greater than or equal to 110°F (43.3°C); and
- c) a Shear Adhesion Failure Temperature (SAFT) of greater than or equal to 140°F (60.0°C); and

wherein the interpolymer comprises a high weight average molecular weight fraction ( $M_{wH}$ ) and a low weight average molecular weight fraction ( $M_{wL}$ ), and wherein the ratio,  $M_{wH}/M_{wL}$ , is from about 1 to about 20, and

wherein the high weight average molecular weight fraction and the low weight average molecular weight fraction are prepared from different catalysts, but the same monomers, under substantially the same polymerization conditions.

- 25 2. The composition of Claim 1, wherein the ethylene interpolymer has iii) a density from 0.88 to 1.06 g/cm<sup>3</sup>.
- The composition of Claim 2, wherein the ethylene interpolymer is derived from olefinic comonomer reactants, comprising at least ethylene and styrene, and has iii) a density from 0.931 to 1.06 g/cm<sup>3</sup>.

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- 4. The composition of Claim 2, wherein the ethylene interpolymer has iii) a density from 0.88 to 0.93 g/cm<sup>3</sup>.
  - The composition of Claim 4, wherein the ethylene interpolymer has:
- i) a density of from 0.89 to 0.92 g/cm<sup>3</sup>;
  - ii) a number average molecular weight (Mn) from 1250 to 7,000; and
  - iii) a Brookfield Viscosity (measured at 149°C/300°F) from 1,000 to 6,000 cP (1.0 to 6.0 Pa·s);

and wherein, when the one or more tackifiers are added in an amount from 20 to 35 percent by weight (based on the combined weight of interpolymer and tackifier) to said ethylene interpolymer, the resulting composition has:

- a) a Brookfield Viscosity (measured at 177°C/350°F) of from 500 to 1,400 cP (0.5 to 1.4 Pa·s);
- b) a Peel Adhesion Failure Temperature (PAFT) of greater than or equal to 115°F (46.1°C); and
- c) a Shear Adhesion Failure Temperature (SAFT) of greater than or equal to 150°F (65.6°C); and
- d) 100% paper tear from 35 to 140°F (1.7 to 60.0°C).
- 20 6. The composition of Claim 4, wherein the ethylene interpolymer has:
  - i) a density from 0.895 to 0.915 g/cm<sup>3</sup>;
  - ii) a number average molecular weight (Mn) from 1500 to 6,000; and
  - iii) a Brookfield Viscosity (measured at 149°C/300°F) from 1,500 to 5,000 cP (1.5 to 5.0 Pa·s);
- and wherein, when the one or more tackifiers are added in an amount from 20 to 35 percent by weight (based on the combined weight of interpolymer and tackifier) to said ethylene interpolymer, then the resulting composition has:
  - a) a Brookfield Viscosity (measured at 177°C/350°F) of from 750 to 1,200 cP (0.75 to 1.2 Pa·s);
- b) a Peel Adhesion Failure Temperature (PAFT) of greater than or equal to 120°F (48.9°C); and





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- c) a Shear Adhesion Failure Temperature (SAFT) of greater than or equal to 170°F (76.7°C); and
- d) a 100% paper tear from 0 to 140°F (-17.8 to 60.0°C).
- 7. The composition of Claim 4, wherein said interpolymer is a copolymer of ethylene/propylene, ethylene/1-butene, ethylene/4-methyl-1-pentene, ethylene/1-pentene, ethylene/1-hexene or ethylene/1-octene.
- 8. The composition of Claim 1, further comprising one or more compounds
  selected from the group consisting of stabilizers, plasticizers, fillers, antioxidants, preservatives, synergists, dyes and pigments.
  - A process of making an ethylene interpolymer, said process comprising:
    i) contacting one or more olefinic monomers in the presence of at least two catalysts, one having a reactivity ratio r<sub>1</sub><sup>H</sup> and the other a reactivity ratio r<sub>1</sub><sup>L</sup>; and
    ii) effectuating the polymerization of the olefinic monomers in a reactor to obtain an olefin polymer, wherein
- iii) each of r<sub>1</sub><sup>H</sup> and r<sub>1</sub><sup>L</sup> is from 1 to 200, and r<sub>1</sub><sup>H</sup>/r<sub>1</sub><sup>L</sup>, is from 0.03 to 30, and wherein one catalyst is capable of producing a first polymer fraction and the other
  catalyst is capable of producing a second polymer fraction, from the same monomers, and under substantially the same polymerization conditions, and wherein the first polymer fraction is a homogeneous linear or substantially linear ethylene polymer with a molecular weight distribution of about 2.4 or less, and the second polymer fraction is a homogeneous linear or substantially linear ethylene polymer with a molecular weight distribution of about 2.4 or less, and/or,
  - weight  $(M_{wH})$  from the monomers under selected polymerization conditions, and the other catalyst is capable of producing a second polymer fraction, with, relative to the first polymer fraction, a low molecular weight  $(M_{wL})$ , from the same monomers under substantially the same polymerization conditions, and where  $M_{wH}/M_{wL}$  is from about 1 to about 20, and

iv) one catalyst is capable of producing a first polymer fraction with a high molecular

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wherein the first polymer fraction  $(M_{wH})$  is a homogeneous linear or substantially linear ethylene polymer with a molecular weight distribution of about 2.4 or less, and the second polymer fraction  $(M_{wL})$  is a homogeneous linear or substantially linear ethylene polymer with a molecular weight distribution of about 2.4 or less.

- 10. The process of Claim 9 wherein the catalysts are single site catalysts.
- 11. The process of Claim 9 wherein the catalysts are metallocene catalysts.
- 12. The process of Claim 11 wherein at least one of the metallocene catalysts is a constrained geometry catalyst.
- 13. The process of Claim 12 wherein said at least one constrained geometry catalyst
   15 is (C<sub>5</sub>Me<sub>4</sub>SiMe<sub>2</sub>N<sup>1</sup>Bu)Ti(η<sup>4</sup>-1,3-pentadiene).
  - 14. The process of Claim 9 wherein the catalysts are (C<sub>5</sub>Me<sub>4</sub>SiMe<sub>2</sub>N<sup>t</sup>Bu)Ti(η<sup>4</sup>-1,3-pentadiene) and (1H-cyclopenta[1]-phenanthrene-2-yl)dimethyl (t-butylamido) silanetitanium dimethyl.
  - 15. The process of Claim 9 wherein the catalysts are (C<sub>5</sub>Me<sub>4</sub>SiMe<sub>2</sub>N<sup>t</sup>Bu)ZrMe<sub>2</sub> and (C<sub>5</sub>Me<sub>4</sub>SiMe<sub>2</sub>N<sup>t</sup>Bu)Ti(η<sup>4</sup>-1,3-pentadiene).
- 16. The process of Claim 9 wherein the catalysts are [N-(1,1-dimethylethyl)-1,1-dimethyl-1-[1,2,3,4,5-η)-3,4-diphenyl-2,4-cyclopentadienyl-1-yl]silanaminato(2)-κN]-dimethyl-titanium and (C<sub>5</sub>Me<sub>4</sub>SiMe<sub>2</sub>N<sup>t</sup>Bu)Ti(η<sup>4</sup>-1,3-pentadiene).
- 17. The process of Claim 9 wherein the catalysts are [N-(1,1-dimethylethyl)-1,1-dimethyl-1-[1,2,3,4,5-η)-3,4-diphenyl-2,4-cyclopentadienyl-1-yl]silanaminato(2)-κN]-dimethyl-titanium and (1H-cyclopenta[1]-phenanthrene-2-yl)dimethyl (t-butylamido) silanetitanium dimethyl.

- 18. The composition of Claim 1, produced by a process, comprising:
  - a) contacting one or more olefinic monomers in the presence of at least a high molecular weight catalyst having a reactivity ratio  $r_1^H$  and at least a low molecular weight catalyst having a reactivity ratio  $r_1^L$  in a single reactor; and
  - b) effectuating the polymerization of the olefinic monomers in the reactor to obtain an olefin polymer; and
  - c) each of  $r_1^H$  and  $r_1^L$  is about 1 to about 200, and  $r_1^H/r_1^L$ , is between 0.03 to 30; and/or
- d) the high molecular weight catalyst is capable of producing a polymer fraction with a high molecular weight, M<sub>wH</sub>, from the monomers under selected polymerization conditions, and the low molecular weight catalyst is capable of producing a polymer fraction with a low molecular weight, M<sub>wL</sub>, from the same monomers under substantially the same polymerization conditions, where M<sub>wH</sub>/M<sub>wL</sub> is from about 1 to about 20.
  - 19. The composition of Claim 1, wherein the ethylene interpolymer contains the residue of at least two catalysts, a first catalyst having a reactivity ratio  $r_1^H$  and a second catalyst having a reactivity ratio  $r_1^L$ , and wherein each of  $r_1^H$  and  $r_1^L$  independently is a number from 1 to 200, and  $r_1^H/r_1^L$  is a number from 0.03 to 30.
  - 20. The composition of Claim 19, wherein the ratio,  $r_1^H/r_1^L$ , is a number greater than 1.

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